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Comments on Kwok's Paper on Acoustic Attenuation by Neutral Donor Impurity Atoms in Germanium[†]

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Kwok[1] has calculated the acoustic attenuation by neutral donor impurities in germanium caused by elastic and inelastic phonon-scattering processes especially for low frequencies, $\hbar\omega \ll 4\Delta$. It is observed that inelastic phonon-scattering processes mainly contribute to the acoustic attenuation, as the acoustic attenuation is proportional to $\tau_{q\lambda}^{-1}$. On the other hand, the phonon conductivity is proportional to $\tau_{q\lambda}$. The elastic phonon-scattering processes will, therefore, contribute more to the phonon conductivity than the inelastic processes. It is, therefore, relatively difficult to estimate the frequency dependence of the elastic phonon-scattering processes from the calculations of the acoustic attenuation.

In recent calculations, Kwok's Eq. (15) was simplified[2] to obtain the relaxation time for the resonant[3] and nonresonant scattering of phonons. The simplified equation (15) of Ref. 1 can be written as (elastic scattering processes only)

$$\tau_{q\lambda}^{-1} = \alpha_{q\lambda} C_{q\lambda} = \frac{\pi}{2\rho C_\lambda^2}\, \omega_{q\lambda} \sum_{q'\,\lambda'} \frac{\omega_{q\lambda}}{\rho C_{\lambda'}^2}\, \delta(\omega_{q\lambda} - \omega_{q'\lambda'})$$

$$\times \left[f_0(T) \left| \sum_m \left(\frac{\Xi_{0m}^{\lambda'}(\vec{q}\,')\, \Xi_{m0}^{\lambda}(\vec{q})}{4\Delta - \hbar\omega_{q\lambda}} + \frac{\Xi_{0m}^{\lambda}(\vec{q})\, \Xi_{m0}^{\lambda'}(\vec{q}\,')}{4\Delta + \hbar\omega_{q\lambda}} \right) \right|^2 \right.$$

$$\left. + f(T) \sum_{n,n'} \left| \left(\frac{\Xi_{n'0}^{\lambda'}(\vec{q})\, \Xi_{0n}^{\lambda}(\vec{q})}{-4\Delta - \hbar\omega_{q\lambda}} + \frac{\Xi_{n'0}^{\lambda}(\vec{q})\, \Xi_{0n}^{\lambda'}(\vec{q}\,')}{-4\Delta + \hbar\omega_{q\lambda}} \right) \right|^2 \right], \qquad (1)$$

where each term is defined in a fashion similar to Kwok. Here the first term in the absolute value corresponds to the phonon scattering of the singlet state; symbolically it can be written as

(a) $\hbar\omega_{q\lambda}$ + (singlet) ⇌ (int triplet) ⇌ $\hbar\omega_{q'\lambda'}$ + (singlet)

(see Fig. 1 of Ref. 3).
The second term stands for the phonon scattering off the triplet state and can be symbolically expressed as

(b) $\hbar\omega_{q\lambda}$ + (triplet) ⇌ (int singlet) ⇌ $\hbar\omega_{q'\lambda'}$ + (triplet)

(see Fig. 1 of Ref. 3).
After simple mathematics, Eq. (1) reduces to the resonance relaxation time[2] as previously obtained by Griffin and Carruthers.[3]

To discuss the acoustic attenuation for low frequencies, $\hbar\omega_{q\lambda} \ll 4\Delta$, Kwok has shown that those terms dominate which correspond to the condition $\epsilon_m = \epsilon_n$. The author, however, feels that we cannot assume any condition like $\epsilon_m = \epsilon_n$ for the above two elastic phonon-scattering processes defined by (a) and (b). This condition is, however, true for inelastic phonon-scattering processes.[1,2]

If we express Eq. (1) as

$$\tau_{q\lambda}^{-1} = \frac{1}{4\pi\rho^2 C_\lambda^2}\, \omega_{q\lambda}^4 F^2(q) \sum_{\lambda'} \frac{F^2[(C_\lambda/C_{\lambda'})q]}{C_{\lambda'}^5}$$

$$\times \left[f_0(T) \left| \sum_m \left(\frac{\tilde{\Xi}_{0m}^{\lambda'}(\vec{q}\,')\, \tilde{\Xi}_{m0}^{\lambda}(\vec{q})}{4\Delta(1 - \hbar\omega_{q\lambda}/4\Delta)} + \frac{\tilde{\Xi}_{0m}^{\lambda}(\vec{q})\, \tilde{\Xi}_{m0}^{\lambda'}(\vec{q}\,')}{4\Delta(1 + \hbar\omega_{q\lambda}/4\Delta)} \right) \right|^2 \right.$$

$$\left. + f(T) \sum_{n,n'} \left| \left(\frac{\tilde{\Xi}_{n'0}^{\lambda}(\vec{q})\, \tilde{\Xi}_{0n}^{\lambda'}(\vec{q}\,')}{4\Delta(1 - \hbar\omega_{q\lambda}/4\Delta)} + \frac{\tilde{\Xi}_{n'0}^{\lambda'}(\vec{q}\,')\, \tilde{\Xi}_{0n}^{\lambda}(\vec{q})}{4\Delta(1 + \hbar\omega_{q\lambda}/4\Delta)} \right) \right|^2 \right], \qquad (2)$$

we can expand $(1 \pm \hbar\omega_{q\lambda}/4\Delta)^{\pm 1}$ using the Binomial theorem as $\hbar\omega_{q\lambda}/4\Delta \ll 1$. We can also neglect the corrections of the order $\hbar\omega_{q\lambda}/4\Delta$. We[4] obtain (considering angular integration)

$$\tau_{q\lambda}^{-1} = \frac{1}{4\pi\rho^2 C_\lambda^2 (4\Delta)^2}\, \omega_{q\lambda}^4 F^2(q) \sum_{\lambda'} \frac{F^2[(C_\lambda/C_{\lambda'})q]}{C_{\lambda'}^5}$$

$$\times \left[4 f_0(T) \left\langle\!\left\langle \left| \sum_m \tilde{\Xi}_{0m}^{\lambda'}(\vec{q}\,')\, \tilde{\Xi}_{m0}^{\lambda}(\vec{q}) \right|^2 \right\rangle\!\right\rangle \right.$$

$$\left. + f(T) \left\langle\!\left\langle \sum_{n,n'} \left| \tilde{\Xi}_{n'0}^{\lambda}(\vec{q})\, \tilde{\Xi}_{0n}^{\lambda'}(\vec{q}\,') + \tilde{\Xi}_{n'0}^{\lambda'}(\vec{q}\,')\, \tilde{\Xi}_{0n}^{\lambda}(\vec{q}) \right|^2 \right\rangle\!\right\rangle \right]. \qquad (3a)$$

Doing some simple mathematics we can express Eq. (3) in terms of an adjustable parameter H as

$$\tau_{q\lambda}^{-1} = H \omega_{q\lambda}^4 F^4(q) \left[f_0(T) + 2 f(T) \right], \qquad (3b)$$

where

$$H = 10 \frac{(\frac{1}{3}E_u)^4}{4\pi\rho^2 C^7 (4\Delta)^2} \left\langle\!\left\langle D_S^{\lambda 1'} \right\rangle\!\right\rangle .$$

Following Kwok, $\tau_{q\lambda}^{-1}$ can be written as

$$\tau_{q\lambda}^{-1} = H \omega_{q\lambda}^4 F^4(q) \left[f_0(T) \left(\frac{\hbar\omega_{q\lambda}}{4\Delta} \right)^2 + f(T) \right]. \qquad (3c)$$

In our calculations,[2] we have used Eq. (3c) (simplified Kwok's expression) to explain the phonon conductivity of As-doped Ge sample for $T < T_R$ $(= 4\Delta/6K_B)$. It is observed that the contribution of the ω^2 term is negligibly small. In a recent communication the results of the phonon conductivity of As-doped Ge [4] is calculated by considering the Kazakov–Nagaev[5] theory of heat transfer in solids. These calculations also suggest that one can neglect the ω^2 term of Eq. (3c) without any appreciable change in the inverse of the relaxation time and hence the phonon conductivity. The only difference with or without using the ω^2 term observed is a slight modification in the value of the adjustable parameter H. These calculations clearly suggest that elastic phonon-scattering processes are effectively proportional to the phonon frequency to its fourth power.

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Dielectric Loss in Fluoride Crystals

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The dipole-reorientation model used by Chen and McDonough to explain dielectric relaxation measurements in BaF_2, CaF_2, and SrF_2 is shown to be inconsistent with previous ionic-conductivity and electric-elastic relaxation measurements and the nominal-impurity-doping levels in their samples.

Recently, Chen and McDonough[1] (CM) reported measurements of the dielectric relaxation in BaF_2, CaF_2, and SrF_2 single crystals doped with YF_3. They attributed the observed relaxation to yttrium-impurity–fluorine-interstitial dipole reorientation and analyzed their data using a dipole-association model in the manner described by Lidiard.[2] We wish to point out that this interpretation of the dielectric loss peak is inconsistent with the nominal-doping levels in their samples and with previous anelastic-anelectric relaxation and ionic-conductivity measurements.

In BaF_2 (0.01% YF_3), the maximum dissipation observed by CM at 178 °C, corresponds to a dipole concentration, formed of F^- interstitials in the nearest-neighbor octahedral site to the Y^{+3} ion, of approximately 1.0%. This calculated dipole concentration exceeds the nominal Y^{+3} concentration they reported by 100 times. In dielectric measurements on BaF_2 (0.01% GdF_3), the authors[3] obtained a loss peak with similar relaxation times[4] to those reported by CM. We did not ascribe this peak to a simple dipole relaxation because of the large disparity between our calculated and measured values of $\tan\delta_{\max}$. Furthermore, the theoretical maximum value of the dipole contribution to the dissipation, computed from our observed value of ω_p (which agrees with the values of CM), and conductivity data obtained on the same material,[3] is approximately 1000 times less than the free-carrier contribution at 175 °C. Chemical and spectrographic analysis of specimens we used showed that the residual concentration of oxygen and other unwanted impurities was insufficient to change the conductivity significantly, say by charge compensation or vacancy or impurity conduction. A similar problem was encountered by Southgate[5] who made elastic relaxation measurements on $CaF_2 - YF_3$ and conductivity measurements on the same material. The parameters of the loss peak determined by CM agree with those determined by Southgate, implying that the same loss mechanism has been observed in both the electric and elastic relaxation measurements. He concluded that an unreasonably high impurity concen-